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(71)Applicant : MITSUBISHI CHEMICALS CORP

(72)Inventor : TAKEHARA HIROAKI

YAMAMOTO TAKAHARU

(54) METHOD AND APPARATUS FOR PRODUCING FULLERENES

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a method and an apparatus able to produce economically a fullerene with mass production.

SOLUTION: Fullerenes are produced by combustion and/or pyrolysis of a carbon containing compound in a combustion furnace. A combustion and/or pyrolysis gas in the combustion furnace is rotated to a combustion furnace axis direction.

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(71)出願人　000005968
三菱化学株式会社
東京都千代田区丸の内二丁目５番２号
(72)発明者　武原　弘明
福岡県北九州市八幡西区黒崎城石１番１号
三菱化学株式会社内
(72)発明者　山本　隆晴
福岡県北九州市八幡西区黒崎城石１番１号
三菱化学株式会社内
(74)代理人　100103997
弁理士　長谷川　曉司

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(54)【発明の名称】　フラーレン類の製造方法およびフラーレン類の製造装置

(57)【要約】
【課題】　経済的かつ大量生産可能なフラーレン製造方法および製造装置を提供する。
【解決する手段】　燃焼炉内において炭素含有化合物を燃焼及び／または熱分解させてフラーレン類を生成するフラーレン類の製造方法であって、燃焼炉内の燃焼及び／または熱分解ガス流を燃焼炉軸方向に対する旋回流とすることを特徴とするフラーレン類の製造方法。

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【特許請求の範囲】

【請求項１】 燃焼炉内において炭素含有化合物を燃焼及び／または熱分解させてフラーレン類を生成するフラーレン類の製造方法であって、燃焼炉内の燃焼及び／または熱分解ガス流を燃焼炉軸方向に対する旋回流とすることを特徴とするフラーレン類の製造方法。

【請求項２】 反応炉内の圧力が大気圧未満である事を特徴とする請求項１に記載のフラーレン類の製造方法。

【請求項３】 燃焼炉内に炭素含有化合物及び／または酸素含有ガスの供給口を有し、各供給口が、各供給口より供給されるガスによって燃焼及び／または熱分解ガス流が反応炉の中心軸に対して旋回流を形成する様に配設されているフラーレン類の製造装置を用いることを特徴とする請求項１または２に記載のフラーレン類の製造方法。

【請求項４】 燃焼炉内に少なくとも炭素含有化合物と酸素含有ガスを供給し、炭素含有化合物を燃焼及び／または熱分解させてフラーレン類を生成するフラーレン類の製造装置において、燃焼炉内の炭素含有化合物及び／または酸素含有ガスの供給口が、各供給口から供給されるガスによって燃焼及び／または熱分解ガス流が反応炉の中心軸に対して旋回流を形成する様に配設されていることを特徴とするフラーレン類の製造装置。

【発明の詳細な説明】

【０００１】

【発明の属する技術分野】 本発明は、フラーレン類の製造方法及びその製造装置に関する。

【０００２】

【従来の技術】 フラーレン類（以下、単にフラーレンと言うことがある。）は、ダイヤモンド、黒鉛に次ぐ第三の炭素同素体の総称であり、C_{60}、C_{70}、などに代表されるように５員環と６員環のネットワークで閉じた中空殻状の炭素分子である。フラーレンの存在が最終的に確認されたのは比較的最近の１９９０年のことであり、比較的新しい炭素材料であるが、その特殊な分子構造ゆえに特異的な物理的性質を示すことが認められ、例えば以下のような広範囲な分野に渡り、革新的な用途開発が急速に展開されつつある。

（１）超硬材料への応用：フラーレンを前駆体とすることで微細結晶粒子をもつ人工ダイヤモンドの精製が可能なため、付加価値のある耐摩耗材料への利用が期待されている。

（２）医薬品への応用：C_{60}誘導体、光デバイスを用いることで抗癌剤、エイズ・骨粗鬆症・アルツハイマー治療薬、造影剤、ステント材料等の用途としての研究が進められている。

（３）超伝導材料への応用：フラーレン薄膜に金属カリウムをドープすると１８Ｋという高い転移温度を持つ超伝導材料をつくり出すことができることが発見され、多方面から注目を集めている。

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（４）半導体製造への応用：レジストにC_{60}を混ぜることでレジスト精度がより一層強化されることを利用し、次世代半導体製造への応用が期待されている。

【０００３】各種炭素数のフラーレンの中でもC_{60}、およびC_{70}は比較的合成が容易であり、それゆえ今後の需要も爆発的に高まることが予想されている。現在知られているフラーレンの製造方法としては以下に示す方法が挙げられる。

（１） レーザー蒸着法：希ガス中に置かれた炭素ターゲットに高エネルギー密度のパルスレーザーを照射し、炭素原子の蒸発により合成する方法。希ガスが流れる石英管を電気炉の中に置き、グラファイト試料をその石英管の中に置く。ガスの流れの上流側からグラファイト試料にレーザーを照射し、蒸発させると電気炉出口付近の冷えた石英管の内壁にC_{60}やC_{70}などのフラーレンを含む煤が付着する。ショット当たりの蒸発量がわずかであり、大量製造には不向き。

（２） 抵抗加熱法：ヘリウムガスで満たされた真空の容器の中でグラファイト棒を通電加熱し昇華させる方法。回路での電気抵抗ロスが大きいので大量製造に不向き。

（３）アーク放電法：数十ｋＰａ中のヘリウムガス中で２本のグラファイト電極を軽く接触させたり、あるいは１～２ｍｍ程度離した状態でアーク放電を起こし、陽極の炭素を昇華させる方法。現在工場規模での大量製造に用いられている。

（４）高周波誘導加熱法：抵抗加熱やアーク放電を使う代わりに、高周波誘導により原料グラファイトに渦電流を流し、これを加熱・蒸発する方法。

（５） 燃焼法：ヘリウム等の不活性ガスと酸素との混合ガス中でベンゼン等の炭化水素原料を不完全燃焼させる方法。ベンゼン燃料の数%が煤となり、その１０%程度がフラーレンとなる点で製造効率は良くないが、複製する煤（フラーレン等）を液体燃料等に使用可能なこと、製造装置が単純である点で、アーク合成法に対抗する大量生産法として注目されている。

（６）ナフタレン熱分解法：ナフタレンを約１０００℃で熱分解させる方法。このように現在までにさまざまなフラーレンの合成法が提案されているが、いずれの方法によってもこれまでにフラーレンを安価に大量に製造する方法は確立されていない。

【０００４】これらの方法のうち、最も安価で、効率的な製造方法の一つと考えられるのは燃焼法であり、特表平６－５０７８７９号公報には、炭素含有物を火炎中で燃焼させ凝縮物を収集することによるフラーレンの製造方法が記載されている。フラーレンはフラーレン等のすす状物質中に含まれて生成されるが、このすす状物質中にフラーレンが含まれる割合をいかに高めるかが大きな課題となっている。

【０００５】また同公報にはフラーレンの収率を向上さ

せるために、火炎温度を上昇させる事．またその手段として外部エネルギー源から火炎にさらにエネルギーを供給する方法が述べられている。好ましいエネルギー源としては、入力流の高気抵抗加熱、マイクロウェーブ加熱．放電加熱及び向流加熱が挙げられている。また一般的にフラーレンの製造は減圧下で行われ、反応領域中に希釈剤を導入する場合もある。これらの減圧度．希釈剤濃度は上記フラーレンの収率に影響を及ぼす事が知られている。

【０００６】そして同公報には、フラーレンの収率および組成は火炎中の滞留時間に依存して変化する　と記載されており、フラーレンを燃焼法で製造する場合．火炎中の滞留時間を均一に保つことがフラーレンの収率を上げ．組成を一定にすることにつながる。一般的に．閉じられた容器の中に火炎を形成させると．火炎中心部と火炎以外の部分で流速差が生じ、燃焼反応が活発に行われる火炎中心部の流速が速くなる。このため、火炎外周部で上流からの燃焼ガスの逆流・巻き込みが起こり．自己循環が発生する場合が多い。

【０００７】このような排ガス自己循環は、火炎温度の局所的高温化を防ぎ、ＮＯｘの発生を抑制する効果がある一方、フラーレンの生成過程において滞留時間の不均一化をもたらす。つまり．自己循環が発生すると．火炎中でフラーレンが生成している段階において、この循環ガスの流れにのったフラーレン前駆体は滞留時間が長くなり．循環ガスの流れにのらないフラーレン前駆体は滞留時間が短くなる。

【０００８】フラーレンは次世代を担う新材料．新素材として多方面から注目されており、このようなフラーレンの滞留時間を制御し、フラーレンを大量に且つ安価に．そして容易に製造する技術の開発が望まれている。

【０００９】

【発明が解決しようとする課題】本発明は前述した様な事情に鑑みてなされたものであり、燃焼方法によるフラーレンの製造において、燃焼炉等の中でのフラーレン前駆体およびフラーレンの滞留時間を制御し、フラーレンを大量に且つ安価に、そして容易に製造する方法を提供することを目的とする。

【００１０】

【課題を解決するための手段】本発明者らは、燃焼方法によるフラーレンの大量且つ安価な製造方法に於いて、最適な燃焼方法を種々検討した結果．フラーレン製造炉内のガス流を旋回流とすることで、火炎中のフラーレンの滞留時間を制御する事ができることを見出し．本発明を完成させた。

【００１１】即ち本発明の要旨は、燃焼炉内において炭素含有化合物を燃焼及び／または熱分解させてフラーレン類を生成するフラーレン類の製造方法であって．燃焼炉内の燃焼及び／または熱分解ガス流を燃焼炉軸方向に対する旋回流とすること．好ましくは反応炉内の圧力を大気圧未満とする、フラーレン類の製造方法に存する。

【００１２】また本発明の今ひとつの要旨は、燃焼炉内に少なくとも炭素含有化合物と酸素含有ガスを供給し、炭素含有化合物を燃焼及び／または熱分解させてフラーレン類を生成するフラーレン類の製造装置において、燃焼炉内の炭素含有化合物及び／または酸素含有ガスの供給口が、各供給口から供給されるガスによって燃焼及び／または熱分解ガス流が反応炉の中心軸に対して旋回流を形成する様に配設されていることを特徴とするフラーレン類の製造装置に存する。

【００１３】

【発明の実施の形態】以下．本発明を具体的に説明する。本発明に係るフラーレンの製造方法及び製造装置について、図１及び図２を用いて説明する。図１は本発明に係るフラーレン製造装置の一例の全体概略断面図であり．図２は酸化ガス導入用ノズルと燃料導入ノズル、原料導入ノズルの配置説明図である。

【００１４】本発明の製造方法においては、燃料及び酸素含有ガス、およびフラーレンの原料である炭素含有化合物（以下単に．原料と言うことがある。）を．（２）（３）（４）の任意の供給口からから供給する。そして燃料を燃焼させる事で火炎を形成し．この中で原料を燃焼させてフラーレンを生成させるか．及び／または燃料の燃焼で形成された燃焼流に原料を接触させることで熱分解によってフラーレンを生成させる。

【００１５】燃料及び酸素含有ガスは．炉内に入る前に混合する、いわゆる予混合燃焼であっても、それぞれ独立したノズルから炉内に供給する、いわゆる拡散燃焼であっても良い。拡散燃焼の場合は、図２において．例えば．中央のノズル（４）から燃料及び原料を供給し、その周囲のノズル（２）（３）から酸素含有ガスを供給する。また、予混合燃焼と拡散燃焼を組み合わせても良く．例えば、図２において．ノズル（２）からは．燃料と酸素含有ガスをあらかじめ混合させたものを導入し、ノズル（３）から酸素含有ガスを、ノズル（４）から原料をそれぞれ独立に供給してもよい。

【００１６】例えば、図２において．ノズル（２）からは燃料と酸素含有化合物を混合したガスを供給して燃焼させ．ノズル（３）から原料を、ノズル（４）から原料の一部燃焼用の酸素含有ガスを供給しても良い。いずれにしても、燃料．原料、酸素含有ガスの供給ノズルは任意であり、燃焼炉内に旋回流を形成することが出来れば．あらゆる組み合わせで供給する事ができる。また、これらの供給口は可動式にしてもよく．炉内の旋回流の状態を調整できるようにする構造が好ましい。

【００１７】図１に示すフラーレン製造装置においては．燃料、原料及び酸素含有ガスはあらかじめ混合され炉内に供給される。燃料炭化水素及び原料炭化水素としては．水素、一酸化炭素．天然ガス．石油ガス等の燃料ガス．重油などの石油系液体燃料、クレオソート油など

の石炭系液体燃料を使用することが出来る。中でもこれらを精製した芳香族系炭化水素を用いることが好ましく、特にベンゼンやトルエン等の芳香族系炭化水素が好ましい。原料の純度は高い方が好ましく、中でも芳香族系炭化水素を用いる際には純度が１００％に近いほど良い。

【００１８】またフラーレンの収率を上げるためには、燃焼過程において希ガス等を用いて希釈する事が好ましい。希ガスは、供給用の専用ノズルから供給しても良いし、燃料、原料、酸素含有ガス中にあらかじめ混合させておいても良い。燃料・原料・酸素含有ガス供給口（２）（３）（４）は、反応炉の同一側に開口している。反応炉内に開口している各供給口の形状は任意であり、略円形、楕円状、三角・四角状などの多角形状やひょうたん型などの不定形であってもよい。

【００１９】燃料・原料・酸素含有ガス供給口の配置は、反応炉に開口していれば任意である。燃料・原料・酸素含有ガス供給口から反応炉内に供給される燃料・原料・酸素含有ガス流は、各供給口が配置されている炉底面に対して任意の角度で供給してよいが、中でも略垂直に配置した上で、炉内部の燃焼ガスが旋回成分を持つように配置する事が重要である。

【００２０】炉内部の燃料及び／または熱分解ガス流が旋回流となるように、各ノズルが配設されている。例えば供給口の一部または全部が、炉の同一平面上に設けられており、且つ炉の中心軸線に対して一定の角度を有するように配置する場合が挙げられる。また図２に示すように、一部のノズルをそのようにし、残りのノズルは炉底面に対して略垂直であっても良い。

【００２１】これら、ノズルを傾ける事により、炉内部に旋回流が発生し、炉内部での排ガス自己循環作用を抑制し、滞留時間を均一に保つ事ができる。また、旋回が＊ない場合に比べて、旋回成分がある分、滞留時間を長く保つ事ができるため、炉の大きさをコンパクトにする事ができる。加えて、例えば、図２におけるノズル（３）とノズル（４）から導入されるガスの流量比率を変更する事で、旋回の強度を制御する事ができ、炉内の滞留時間を任意に変更する事ができる。

【００２２】これにより、得られるフラーレンの収率を最大に調整する事ができるとともに、フラーレンの種類を制御することもできる。反応炉内に供給される酸素含有ガス流および燃料流の流速は適宜選択すると共に反応炉内の温度変化などに応じて調整すればよい。炉内の燃焼温度も重要で、少なくとも１０００℃以上、中でも１４００℃以上、特に１６００℃以上とするのが好ましい。

【００２３】炉内圧力は減圧状態とし、大気圧未満であることが好ましく、より好ましい範囲は１０～３００［ｔｏｒｒ］である。燃料の希釈剤濃度は、実質的に０～４０モル％の範囲であり、また酸素含有ガスの酸素以外の成分（希釈剤）濃度は０～９０％モルの範囲で任意に調整できる。

【００２４】

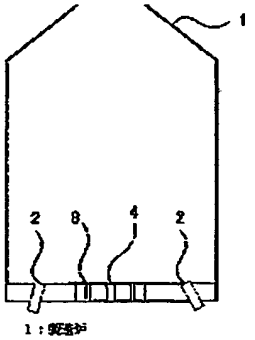
【発明の効果】本発明の製造方法によって、経済的な燃焼方法によるフラーレンの大量且つ安価な製造方法を提供することが出来、さらに燃焼方法においてフラーレン製造炉内のガス流を旋回流とすることでフラーレンの製造条件を調整することが出来る。

【図面の簡単な説明】

【図１】　図１は本発明に係るフラーレン製造装置の一例の全体概略断面図である。

【図２】　図２は酸化ガス導入用ノズルと燃料導入ノズル、原料導入ノズルの配置説明図である。

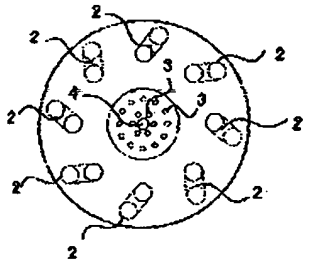
【図１】



１：製造炉

２，３，４：燃料、原料、酸素含有ガス供給口

【図２】



２，３，４：燃料、原料、酸素含有ガス供給口

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CLAIMS

[Claim(s)]
[Claim 1] The manufacture approach of the fullerene characterized by being the manufacture approach of combustion and/or the fullerene which are made to carry out a pyrolysis and generate fullerene, and making a carbon content compound into a turning style [as opposed to combustion furnace shaft orientations for the combustion in a combustion furnace, and/or a cracked gas style] into a combustion furnace.
[Claim 2] The manufacture approach of the fullerene according to claim 1 characterized by the pressure in a reactor being under atmospheric pressure.
[Claim 3] The manufacture approach of the fullerene according to claim 1 or 2 characterized by using the manufacturing installation of the fullerene currently arranged so that combustion and/or a cracked gas style may form a turning style to the medial axis of a reactor by the gas by which it has the feed hopper of a carbon content compound and/or oxygen content gas, and each feed hopper is supplied from each feed hopper in a combustion furnace.
[Claim 4] The manufacturing installation of the fullerene characterized by to be arranged so that combustion and/or a cracked-gas style may form a turning style to the medial axis of a reactor by the gas by which a carbon content compound and oxygen content gas are supplied at least in a combustion furnace, and a carbon content compound is supplied to the feed hopper of the carbon content compound in a combustion furnace, and/or oxygen content gas from each feed hopper in the manufacturing installation of combustion and/or the fullerene which are made to carry out a pyrolysis and generate fullerene.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention] This invention relates to the manufacture approach of fullerene, and its manufacturing installation.
[0002]
[Description of the Prior Art] Fullerene (it may only be hereafter called fullerene) is the generic names of the third carbon allotrope which ranks second to a diamond and a graphite, and it is the carbon molecule of the shape of hollow husks closed in the network of five membered-rings and six membered-rings as represented in C60, C70, etc. Although it is comparatively that existence of fullerene was checked eventually and it is a comparatively new carbon material, it is admitted that the special molecular structure, therefore specific physical property are shown, for example, innovative application development is being quickly developed over the wide range following fields.
(1) Application to a superhard ingredient : since purification of the artificial diamond which has a fine crystal grain child by using fullerene as a precursor is possible, utilization to an abrasion resistance material with added value is expected.
(2) Application to drugs : research as an application of an anticancer agent, an acquired immunodeficiency syndrome, osteoporosis and the Alzheimer remedy, a contrast medium, a stent ingredient, etc. is advanced by using C60 derivative and an optical device.
(3) Application to a superconducting material : if metallic potassium is doped to a fullerene thin film, it is discovered that a superconducting material with a high transition temperature called 18K can be made, and since various, attract attention.
(4) Application to semi-conductor manufacture : it uses that resist structure is further strengthened with mixing C60 with a resist, and the application to next-generation semi-conductor manufacture is expected.
[0003] Also in the fullerene of various carbon numbers, C60 and C70 are comparatively easy to compound, and it is expected that future need so also increases explosively. The approach shown below as the manufacture approach of fullerene learned now is mentioned.
(1) Laser vacuum deposition : how to irradiate the pulse laser of a high energy consistency at the carbon target placed into rare gas, and compound by evaporation of a carbon atom. The quartz tube with which rare gas flows is placed into an electric furnace, and a graphite sample is placed into the quartz tube. If laser is irradiated and is evaporated in a graphite sample from the upstream of the flow of gas, the soot containing fullerene, such as C60 and C70, will adhere to the wall of the quartz tube with which near the electric furnace outlet got cold. The evaporation per shot is slight and unsuitable for large quantity manufacture.
(2) Resistance heating method : the approach to which carry out energization heating and a graphite rod is made to sublimate in the container of the vacuum filled with gaseous helium. Unsuitable for large quantity manufacture, since the electric resistance loss in a circuit is large.
(3) Arc discharge method : the approach to which the carbon of a lifting and an anode plate is made to

sublimate arc discharge in the condition of having contacted two graphite electrodes lightly in the gaseous helium in dozens kPa(s), or having detached about 1-2mm. It is used for large quantity manufacture on current works magnitude.
(4) Radio frequency heating method : how to heat and evaporate a sink and this in an eddy current by RF induction at raw material graphite instead of using resistance heating and arc discharge.
(5) Combustion method : the approach of carrying out the incomplete combustion of the hydrocarbon raw materials, such as benzene, in the mixed gas of inert gas, such as helium, and oxygen. It is observed as the mass-producing method for being usable to liquid fuel etc., and the point that a manufacturing installation is simple, and opposing an arc synthesis method in the soot (fullerene etc.) reproduced in that several% of a benzene fuel serves as soot, and the about 10% becomes fullerene although manufacture effectiveness is not good.
(6) Naphthalene thermal decomposition method : the approach of carrying out the pyrolysis of the naphthalene at about 1000 degrees C. Thus, although the synthesis method of various fullerene by current is proposed, the method of manufacturing fullerene to a large quantity cheaply by any approach until now is not established.
[0004] A combustion method is considered one of these approaches of the cheapest and efficient manufacture approach, and the manufacture approach of the fullerene by burning a carbon inclusion in a flame in a ****** No. 507879 [six to] official report, and collecting condensates in it is indicated. Although fullerene is contained in soot-like matter, such as fullerene, and it is generated, it has been a big technical problem how the rate that fullerene is contained in this soot-like matter is raised.
[0005] Moreover, in order to raise the yield of fullerene in this official report, the approach of supplying energy further is stated to the flame from the external energy source as raising flame temperature and its means. As a desirable energy source, electric resistance heating of an input style, microwave heating, discharge heating, and counterflow heating are mentioned. Moreover, generally, manufacture of fullerene is performed under reduced pressure and a diluent may be introduced all over a reaction field. It is known whenever [these reduced pressure] that diluent concentration will affect the yield of the above-mentioned fullerene.
[0006] And the yield of fullerene and a presentation change to this official report depending on the residence time in a flame. When it is indicated and manufactures fullerene with a combustion method, it leads to maintaining the residence time in a flame at homogeneity carrying out yield of fullerene raising, and carrying out a presentation to regularity. If a flame is made to form into the closed container generally, the rate-of-flow difference will arise in parts other than a flame core and a flame, and the rate of flow of the flame core where a combustion reaction is performed actively will become quick. For this reason, a back run and contamination of the combustion gas from the upstream happen in the flame periphery section, and self-circulation occurs in many cases.
[0007] Such exhaust gas self-circulation prevents local elevated-temperature-ization of flame temperature, and while there is effectiveness which controls generating of NOx, it brings about ununiformity-ization of the residence time in the generation process of fullerene. That is, as for the fullerene precursor with which, as for the fullerene precursor which rode the flow of this circulating gas in the phase which fullerene is generating in the flame, the residence time does not ride the flow of circulating gas by becoming long, the residence time will become short if self-circulation occurs.
[0008] Since fullerene is various as the exotic material which bears the next generation, and new materials, it is observed, and the residence time of such fullerene is controlled, and development of the technique of manufacturing fullerene cheaply and easily in large quantities is desired.
[0009]
[Problem(s) to be Solved by the Invention] This invention is made in view of a situation which was mentioned above, the fullerene precursor in the inside of a combustion furnace etc. and the residence time of BURAREN are controlled in manufacture of the fullerene by the combustion method, and it aims at offering the approach of manufacturing fullerene cheaply and easily in large quantities.
[0010]
[Means for Solving the Problem] In the large quantity and the cheap manufacture approach of fullerene

by the combustion method, as a result of examining various optimal combustion methods, this invention persons are making the gas stream in a fullerene manufacture furnace into a turning style, and completed a header and this invention for the residence time of the fullerene in a flame being controllable.
[0011] That is, the summary of this invention consists in the manufacture approach of being combustion and/or the manufacture approach of fullerene which is made to carry out a pyrolysis and generates fullerene, and making a carbon content compound into a turning style [as opposed to combustion furnace shaft orientations for the combustion in a combustion furnace, and/or a cracked gas style] into a combustion furnace, and the fullerene which make the pressure in a reactor under atmospheric pressure preferably.
[0012] Moreover, the unsatisfactory summary of this invention supplies a carbon content compound and oxygen content gas at least in a combustion furnace, and a carbon content compound is set to the manufacturing installation of combustion and/or the fullerene which are made to carry out a pyrolysis and generate fullerene. The feed hopper of the carbon content compound in a combustion furnace and/or oxygen content gas consists in the manufacturing installation of the fullerene characterized by being arranged so that combustion and/or a cracked gas style may form a turning style to the medial axis of a reactor by the gas supplied from each feed hopper.
[0013]
[Embodiment of the Invention] Hereafter, this invention is explained concretely. The manufacture approach of fullerene and manufacturing installation concerning this invention are explained using drawing 1 and drawing 2 . Drawing 1 is the whole example outline sectional view of the fullerene manufacturing installation concerning this invention, and drawing 2 is the arrangement explanatory view of the nozzle for oxidation gas installation, a fuel installation nozzle, and a raw material installation nozzle.
[0014] the carbon content compound (it may only be called a raw material below) which are a fuel, oxygen content gas, and the raw material of fullerene in the manufacture approach of this invention -- from the feed hopper of the arbitration of (2), (3), and (4) -- since -- it supplies. And a pyrolysis is made to generate fullerene by contacting a raw material in the style of [which formed the flame by burning a fuel, the raw material was burned in this, and was made to generate fullerene, or was formed by combustion of a fuel] combustion.
[0015] Even if a fuel and oxygen content gas are the so-called premixed combustion mixed before entering in a furnace, they may be the so-called diffusive burning supplied in a furnace from the nozzle which became independent, respectively. In drawing 2 , in the case of diffusive burning, a fuel and a raw material are supplied from a central nozzle (4), and it supplies oxygen content gas from the nozzle (2) of the perimeter, and (3). Moreover, premixed combustion and diffusive burning may be combined, for example, in drawing 2 , from a nozzle (2), what mixed oxygen content gas with the fuel beforehand may be introduced, and a nozzle (4) to a raw material may be independently supplied for oxygen content gas from a nozzle (3), respectively.
[0016] for example, in drawing 2 , from a nozzle (2), the gas which mixed the oxygen content compound with the fuel is supplied, and it burns -- making -- a nozzle (3) to a raw material -- a nozzle (4) to some raw materials -- the oxygen content gas for combustion may be supplied. Anyway, the supply nozzle of a fuel, a raw material, and oxygen content gas is arbitrary, and if a turning style can be formed in a combustion furnace, it can be supplied in all combination. Moreover, these feed hoppers have the desirable structure of making it working and enabling it to adjust the condition of the turning style in a furnace.
[0017] In the fullerene manufacturing installation shown in drawing 1 , it is mixed beforehand and a fuel, a raw material, and oxygen content gas are supplied in a furnace. As a fuel hydrocarbon and coal-for-coke-making-ized hydrogen, coal system liquid fuel, such as petroleum system liquid fuel, such as fuel gas, such as hydrogen, a carbon monoxide, natural gas, and petroleum gas, and a fuel oil, and creosote oil, can be used. Especially, it is desirable to use the aromatic series system hydrocarbon which refined these, and aromatic series system hydrocarbons, such as benzene and toluene, are especially desirable. Its higher one is desirable, and it is so good that its purity is close to 100% in case the purity

of a raw material uses an aromatic series system hydrocarbon especially.
[0018] Moreover, in order to gather the yield of fullerene, it is desirable to dilute using rare gas etc. in a combustion process. Rare gas may be supplied from the exclusive nozzle for supply, and may be beforehand mixed in a fuel, a raw material, and oxygen content gas. Opening of a fuel, a raw material and oxygen content gas supply opening (2), (3), and (4) is carried out to the same reactor side. The configuration of each feed hopper which is carrying out opening into the reactor may be arbitrary, and may be the infinite form of the shape of a polygon, such as the shape of an approximate circle form and an ellipse, and the shape of a triangle and a rectangular head, a gourd mold, etc.
[0019] If opening of the arrangement of a fuel, a raw material, and oxygen content gas supply opening is carried out to the reactor, it is arbitrary. Although the fuel, raw material, and oxygen content gas stream supplied in a reactor may be supplied at an angle of arbitration to the blast furnace bottom side where each feed hopper is arranged from a fuel, a raw material, and oxygen content gas supply opening, it is important to arrange so that the combustion gas inside a furnace may have a turning component when it has arranged to the abbreviation perpendicular especially.
[0020] Each nozzle is arranged so that the fuel and/or cracked gas style inside a furnace may turn into a turning style. For example, the case where a part or all of a feed hopper arranges so that it may be prepared on the same flat surface of a furnace and may have a fixed include angle to the medial-axis line of a furnace is mentioned. moreover, it is shown in drawing 2 -- as -- some nozzles -- such -- carrying out -- the remaining nozzles -- a blast furnace bottom side -- receiving -- abbreviation -- it may be vertical.
[0021] By leaning these nozzles, a turning style can occur inside a furnace, the exhaust gas self-circulation operation inside a furnace can be controlled, and the residence time can be maintained at homogeneity. Moreover, since a part and the residence time with a turning component can be kept long compared with the case where there is no turning, magnitude of a furnace can be used as a compact. In addition, the reinforcement of turning can be controlled by, for example, changing the rate of flow rate of the gas introduced from the nozzle (3) and nozzle (4) in drawing 2 , and the residence time in a furnace can be changed into arbitration by it.
[0022] The class of fullerene is also controllable while being able to adjust the yield of the fullerene obtained to max by this. What is necessary is just to adjust the rate of flow of the oxygen content gas stream supplied in a reactor, and a fuel style according to the temperature change in a reactor etc. while choosing suitably. The combustion temperature in a furnace is also important and it is desirable to make 1400 degrees C or more especially into 1600 degrees C or more also in at least 1000 degrees C or more.
[0023] Furnace internal pressure is made into a reduced pressure condition, it is desirable that it is under atmospheric pressure, and more desirable range is 10-300 [torr]. The 0-40-mol range of the diluent concentration of a fuel is % substantially, and component (diluent) concentration other than the oxygen of oxygen content gas can be adjusted to arbitration in the range of a mol 0 to 90%.
[0024]
[Effect of the Invention] The manufacture conditions of fullerene can be adjusted by being able to offer the large quantity and the cheap manufacture approach of fullerene by the economical combustion method, and making the gas stream in a fullerene manufacture furnace into a turning style in a combustion method by the manufacture approach of this invention, further.

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TECHNICAL FIELD

[Field of the Invention] This invention relates to the manufacture approach of fullerene, and its manufacturing installation.

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PRIOR ART

[Description of the Prior Art] Fullerene (it may only be hereafter called fullerene) is the generic names of the third carbon allotrope which ranks second to a diamond and a graphite, and it is the carbon molecule of the shape of hollow husks closed in the network of five membered-rings and six membered-rings as represented in C60, C70, etc. Although it is comparatively that existence of fullerene was checked eventually and it is a comparatively new carbon material, it is admitted that the special molecular structure, therefore specific physical property are shown, for example, innovative application development is being quickly developed over the wide range following fields.
(1) Application to a superhard ingredient : since purification of the artificial diamond which has a fine crystal grain child by using fullerene as a precursor is possible, utilization to an abrasion resistance material with added value is expected.
(2) Application to drugs : research as an application of an anticancer agent, an acquired immunodeficiency syndrome, osteoporosis and the Alzheimer remedy, a contrast medium, a stent ingredient, etc. is advanced by using C60 derivative and an optical device.
(3) Application to a superconducting material : if metallic potassium is doped to a fullerene thin film, it is discovered that a superconducting material with a high transition temperature called 18K can be made, and since various, attract attention.
(4) Application to semi-conductor manufacture : it uses that resist structure is further strengthened with mixing C60 with a resist, and the application to next-generation semi-conductor manufacture is expected.
[0003] Also in the fullerene of various carbon numbers, C60 and C70 are comparatively easy to compound, and it is expected that future need so also increases explosively. The approach shown below as the manufacture approach of fullerene learned now is mentioned.
(1) Laser vacuum deposition : how to irradiate the pulse laser of a high energy consistency at the carbon target placed into rare gas, and compound by evaporation of a carbon atom. The quartz tube with which rare gas flows is placed into an electric furnace, and a graphite sample is placed into the quartz tube. If laser is irradiated and is evaporated in a graphite sample from the upstream of the flow of gas, the soot containing fullerene, such as C60 and C70, will adhere to the wall of the quartz tube with which near the electric furnace outlet got cold. The evaporation per shot is slight and unsuitable for large quantity manufacture.
(2) Resistance heating method : the approach to which carry out energization heating and a graphite rod is made to sublimate in the container of the vacuum filled with gaseous helium. Unsuitable for large quantity manufacture, since the electric resistance loss in a circuit is large.
(3) Arc discharge method : the approach to which the carbon of a lifting and an anode plate is made to sublimate arc discharge in the condition of having contacted two graphite electrodes lightly in the gaseous helium in dozens kPa(s), or having detached about 1-2mm. It is used for large quantity manufacture on current works magnitude.
(4) Radio frequency heating method : how to heat and evaporate a sink and this in an eddy current by RF induction at raw material graphite instead of using resistance heating and arc discharge.

(5) Combustion method : the approach of carrying out the incomplete combustion of the hydrocarbon raw materials, such as benzene, in the mixed gas of inert gas, such as helium, and oxygen. It is observed as the mass-producing method for being usable to liquid fuel etc., and the point that a manufacturing installation is simple, and opposing an arc synthesis method in the soot (fullerene etc.) reproduced in that several% of a benzene fuel serves as soot, and the about 10% becomes fullerene although manufacture effectiveness is not good.
(6) Naphthalene thermal decomposition method : the approach of carrying out the pyrolysis of the naphthalene at about 1000 degrees C. Thus, although the synthesis method of various fullerene by current is proposed, the method of manufacturing fullerene to a large quantity cheaply by any approach until now is not established.
[0004] A combustion method is considered one of these approaches of the cheapest and efficient manufacture approach, and the manufacture approach of the fullerene by burning a carbon inclusion in a flame in a ****** No. 507879 [six to] official report, and collecting condensates in it is indicated. Although fullerene is contained in soot-like matter, such as fullerene, and it is generated, it has been a big technical problem how the rate that fullerene is contained in this soot-like matter is raised.
[0005] Moreover, in order to raise the yield of fullerene in this official report, the approach of supplying energy further is stated to the flame from the external energy source as raising flame temperature and its means. As a desirable energy source, electric resistance heating of an input style, microwave heating, discharge heating, and counterflow heating are mentioned. Moreover, generally, manufacture of fullerene is performed under reduced pressure and a diluent may be introduced all over a reaction field. It is known whenever [these reduced pressure] that diluent concentration will affect the yield of the above-mentioned fullerene.
[0006] And the yield of fullerene and a presentation change to this official report depending on the residence time in a flame. When it is indicated and manufactures fullerene with a combustion method, it leads to maintaining the residence time in a flame at homogeneity carrying out yield of fullerene raising, and carrying out a presentation to regularity. If a flame is made to form into the closed container generally, the rate-of-flow difference will arise in parts other than a flame core and a flame, and the rate of flow of the flame core where a combustion reaction is performed actively will become quick. For this reason, a back run and contamination of the combustion gas from the upstream happen in the flame periphery section, and self-circulation occurs in many cases.
[0007] Such exhaust gas self-circulation prevents local elevated-temperature-ization of flame temperature, and while there is effectiveness which controls generating of NOx, it brings about ununiformity-ization of the residence time in the generation process of fullerene. That is, as for the fullerene precursor with which, as for the fullerene precursor which rode the flow of this circulating gas in the phase which fullerene is generating in the flame, the residence time does not ride the flow of circulating gas by becoming long, the residence time will become short if self-circulation occurs.
[0008] Since fullerene is various as the exotic material which bears the next generation, and new materials, it is observed, and the residence time of such fullerene is controlled, and development of the technique of manufacturing fullerene cheaply and easily in large quantities is desired.

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EFFECT OF THE INVENTION

[Effect of the Invention] The manufacture conditions of fullerene can be adjusted by being able to offer the large quantity and the cheap manufacture approach of fullerene by the economical combustion method, and making the gas stream in a fullerene manufacture furnace into a turning style in a combustion method by the manufacture approach of this invention, further.

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TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] This invention is made in view of a situation which was mentioned above, the fullerene precursor in the inside of a combustion furnace etc. and the residence time of BURAREN are controlled in manufacture of the fullerene by the combustion method, and it aims at offering the approach of manufacturing fullerene cheaply and easily in large quantities.

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MEANS

[Means for Solving the Problem] In the large quantity and the cheap manufacture approach of fullerene by the combustion method, as a result of examining various optimal combustion methods, this invention persons are making the gas stream in a fullerene manufacture furnace into a turning style, and completed a header and this invention for the residence time of the fullerene in a flame being controllable.
[0011] That is, the summary of this invention consists in the manufacture approach of being combustion and/or the manufacture approach of fullerene which is made to carry out a pyrolysis and generates fullerene, and making a carbon content compound into a turning style [as opposed to combustion furnace shaft orientations for the combustion in a combustion furnace, and/or a cracked gas style] into a combustion furnace, and the fullerene which make the pressure in a reactor under atmospheric pressure preferably.
[0012] Moreover, the unsatisfactory summary of this invention supplies a carbon content compound and oxygen content gas at least in a combustion furnace, and a carbon content compound is set to the manufacturing installation of combustion and/or the fullerene which are made to carry out a pyrolysis and generate fullerene. The feed hopper of the carbon content compound in a combustion furnace and/or oxygen content gas consists in the manufacturing installation of the fullerene characterized by being arranged so that combustion and/or a cracked gas style may form a turning style to the medial axis of a reactor by the gas supplied from each feed hopper.
[0013]
[Embodiment of the Invention] Hereafter, this invention is explained concretely. The manufacture approach of fullerene and manufacturing installation concerning this invention are explained using drawing 1 and drawing 2 . Drawing 1 is the whole example outline sectional view of the fullerene manufacturing installation concerning this invention, and drawing 2 is the arrangement explanatory view of the nozzle for oxidation gas installation, a fuel installation nozzle, and a raw material installation nozzle.
[0014] the carbon content compound (it may only be called a raw material below) which are a fuel, oxygen content gas, and the raw material of fullerene in the manufacture approach of this invention -- from the feed hopper of the arbitration of (2), (3), and (4) -- since -- it supplies. And a pyrolysis is made to generate fullerene by contacting a raw material in the style of [which formed the flame by burning a fuel, the raw material was burned in this, and was made to generate fullerene, or was formed by combustion of a fuel] combustion.
[0015] Even if a fuel and oxygen content gas are the so-called premixed combustion mixed before entering in a furnace, they may be the so-called diffusive burning supplied in a furnace from the nozzle which became independent, respectively. In drawing 2 , in the case of diffusive burning, a fuel and a raw material are supplied from a central nozzle (4), and it supplies oxygen content gas from the nozzle (2) of the perimeter, and (3). Moreover, premixed combustion and diffusive burning may be combined, for example, in drawing 2 , from a nozzle (2), what mixed oxygen content gas with the fuel beforehand may be introduced, and a nozzle (4) to a raw material may be independently supplied for oxygen content gas from a nozzle (3), respectively.

[0016] for example, in drawing 2 , from a nozzle (2), the gas which mixed the oxygen content compound with the fuel is supplied, and it burns -- making -- a nozzle (3) to a raw material -- a nozzle (4) to some raw materials -- the oxygen content gas for combustion may be supplied. Anyway, the supply nozzle of a fuel, a raw material, and oxygen content gas is arbitrary, and if a turning style can be formed in a combustion furnace, it can be supplied in all combination. Moreover, these feed hoppers have the desirable structure of making it working and enabling it to adjust the condition of the turning style in a furnace.

[0017] In the fullerene manufacturing installation shown in drawing 1 , it is mixed beforehand and a fuel, a raw material, and oxygen content gas are supplied in a furnace. As a fuel hydrocarbon and coal-for-coke-making-ized hydrogen, coal system liquid fuel, such as petroleum system liquid fuel, such as fuel gas, such as hydrogen, a carbon monoxide, natural gas, and petroleum gas, and a fuel oil, and creosote oil, can be used. Especially, it is desirable to use the aromatic series system hydrocarbon which refined these, and aromatic series system hydrocarbons, such as benzene and toluene, are especially desirable. Its higher one is desirable, and it is so good that its purity is close to 100% in case the purity of a raw material uses an aromatic series system hydrocarbon especially.

[0018] Moreover, in order to gather the yield of fullerene, it is desirable to dilute using rare gas etc. in a combustion process. Rare gas may be supplied from the exclusive nozzle for supply, and may be beforehand mixed in a fuel, a raw material, and oxygen content gas. Opening of a fuel, a raw material and oxygen content gas supply opening (2), (3), and (4) is carried out to the same reactor side. The configuration of each feed hopper which is carrying out opening into the reactor may be arbitrary, and may be the infinite form of the shape of a polygon, such as the shape of an approximate circle form and an ellipse, and the shape of a triangle and a rectangular head, a gourd mold, etc.

[0019] If opening of the arrangement of a fuel, a raw material, and oxygen content gas supply opening is carried out to the reactor, it is arbitrary. Although the fuel, raw material, and oxygen content gas stream supplied in a reactor may be supplied at an angle of arbitration to the blast furnace bottom side where each feed hopper is arranged from a fuel, a raw material, and oxygen content gas supply opening, it is important to arrange so that the combustion gas inside a furnace may have a turning component when it has arranged to the abbreviation perpendicular especially.

[0020] Each nozzle is arranged so that the fuel and/or cracked gas style inside a furnace may turn into a turning style. For example, the case where a part or all of a feed hopper arranges so that it may be prepared on the same flat surface of a furnace and may have a fixed include angle to the medial-axis line of a furnace is mentioned. moreover, it is shown in drawing 2 -- as -- some nozzles -- such -- carrying out -- the remaining nozzles -- a blast furnace bottom side -- receiving -- abbreviation -- it may be vertical.

[0021] By leaning these nozzles, a turning style can occur inside a furnace, the exhaust gas self-circulation operation inside a furnace can be controlled, and the residence time can be maintained at homogeneity. Moreover, since a part and the residence time with a turning component can be kept long compared with the case where there is no turning, magnitude of a furnace can be used as a compact. In addition, the reinforcement of turning can be controlled by, for example, changing the rate of flow rate of the gas introduced from the nozzle (3) and nozzle (4) in drawing 2 , and the residence time in a furnace can be changed into arbitration by it.

[0022] The class of fullerene is also controllable while being able to adjust the yield of the fullerene obtained to max by this. What is necessary is just to adjust the rate of flow of the oxygen content gas stream supplied in a reactor, and a fuel style according to the temperature change in a reactor etc. while choosing suitably. The combustion temperature in a furnace is also important and it is desirable to make 1400 degrees C or more especially into 1600 degrees C or more also in at least 1000 degrees C or more.

[0023] Furnace internal pressure is made into a reduced pressure condition, it is desirable that it is under atmospheric pressure, and more desirable range is 10-300 [torr]. The 0-40-mol range of the diluent concentration of a fuel is % substantially, and component (diluent) concentration other than the oxygen of oxygen content gas can be adjusted to arbitration in the range of a mol 0 to 90%.

[Translation done.]

* NOTICES *

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]
[Drawing 1] Drawing 1 is the whole example outline sectional view of the fullerene manufacturing installation concerning this invention.
[Drawing 2] Drawing 2 is the arrangement explanatory view of the nozzle for oxidation gas installation, a fuel installation nozzle, and a raw material installation nozzle.

[Translation done.]

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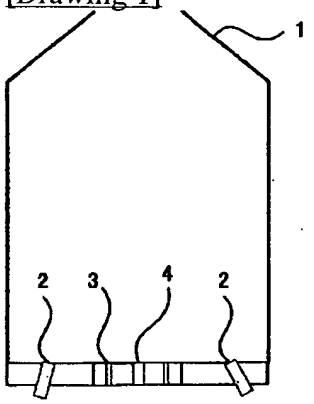
DRAWINGS

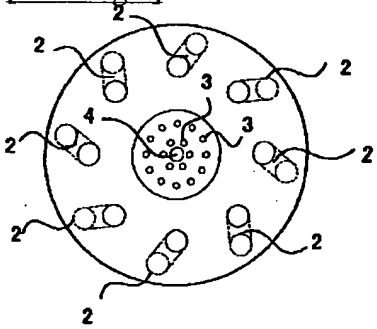
[Drawing 1]



1：製造炉

2，3，4：燃料、原料、酸素含有ガス供給口

[Drawing 2]



2，3，4：燃料、原料、酸素含有ガス供給口．

[Translation done.]